First Principles Study of Superconductivity in FeSe

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Outline

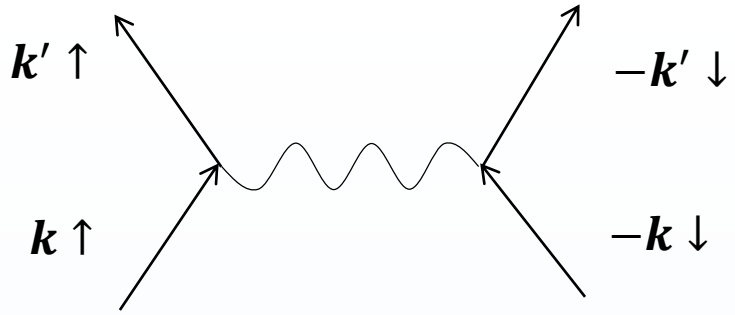
- Superconductivity
- Iron-based Superconductors
  - Typical iron-based superconductors
  - Structural and magnetic properties of FeSe
- Calculation Results of FeSe
- Conclusion

Superconductivity

- First discovered by H. K. Onnes[1] (1911)
  - Mercury with zero resistivity when $T < 4.2$ K
- Properties when $T < T_C$
  - Zero resistance
  - Expels magnetic fields, leaving no magnetic field inside (Meissner effect)

BCS Theory

- Electron-Phonon Interaction:
  - Cooper Pairs[1]
  - Condition: $\epsilon_F - \hbar\omega_D < \epsilon_{\boldsymbol{k}} < \epsilon_F + \hbar\omega_D$[2]



- BCS Equation:
  - $kT_c = 1.14\hbar\omega_D \exp[-\frac{1}{D(\epsilon_F)V}]$[2]

[1]: L. N. Cooper, Phys. Rev. 104, 1189 (1956)
[2]: Bardeen, Cooper and Schrieffer, Phys. Rev. 108, 1175 (1957)

McMillan's formula

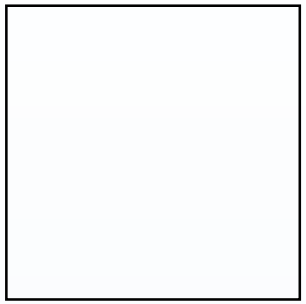
- Electron-Phonon Coupling Strength:[1]
  - $\lambda = 2\int_0^\infty \frac{1}{\omega}[\alpha^2 F(\omega)]d\omega$

- McMillan's formula:[1]
  - $T_c = \frac{\omega_{log}}{1.2}\exp[\frac{-1.04(1+\lambda)}{\lambda-\mu^*(1+0.62\lambda)}]$
    - $\mu^*: 0.10\sim0.16$

[1]: W. L. McMillan, PhysRev.167.331 (1968)

Typical Iron-based Superconductors

- Tetragonal-orthorhombic structural transition at T_S



4-fold symmetry

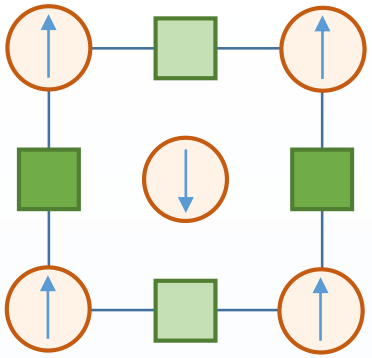
Top view of tetragonal structure

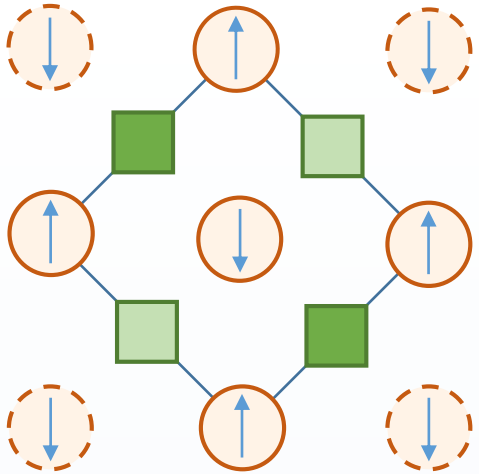
2-fold symmetry

Top view of orthorhombic structure

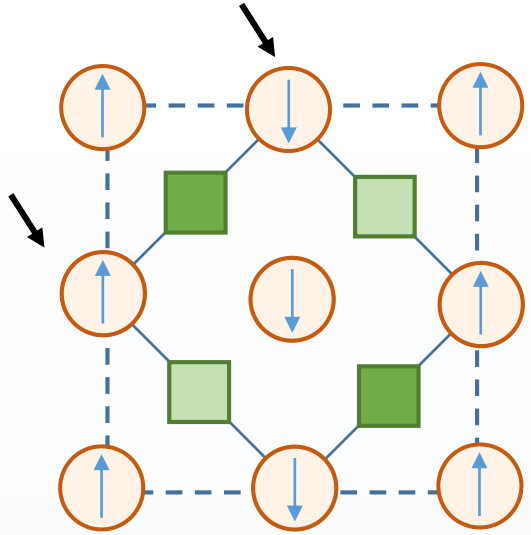
- stripe-AFM transition at T_N
  - Symmetry breaking

Stripe Anti-Ferromagnetic State





Checkerboard AFM

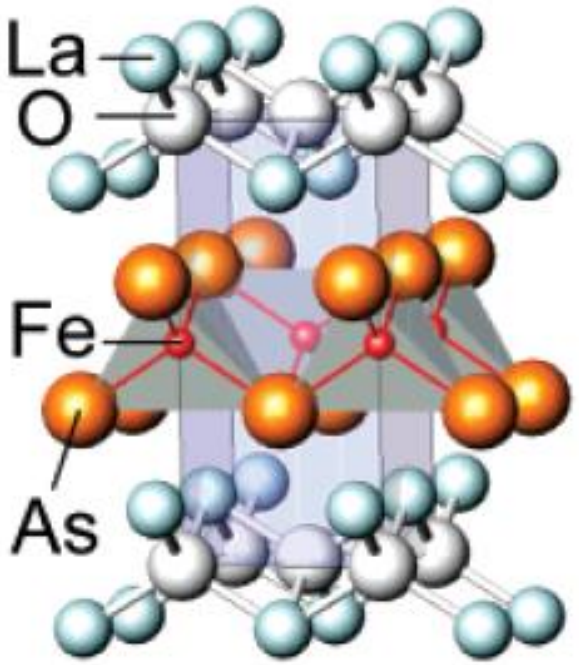


Stripe AFM
Symmetry breaking
$(\sqrt{2} \times \sqrt{2})$ super-cell

Example: $La(O_{1-x}F_x)FeAs$

- Undoped LaOFeAs:
  - Structure transforms from tetragonal (*P4/nmm*) to orthorhombic (*Cmme*) below T_S (150 K) [1]
  - Magnetism transforms to stripe-AFM below T_N (134 K) [1]
  - No superconductivity
- Doped $La(O_{1-x}F_x)FeAs$: (x = 0.05-0.12)
  - Tetragonal structure (*P4/nmm*). No T_S and T_N.
  - Superconductivity occurs below T_C (26 K) [2]

Structure of LaOFeAs [2]

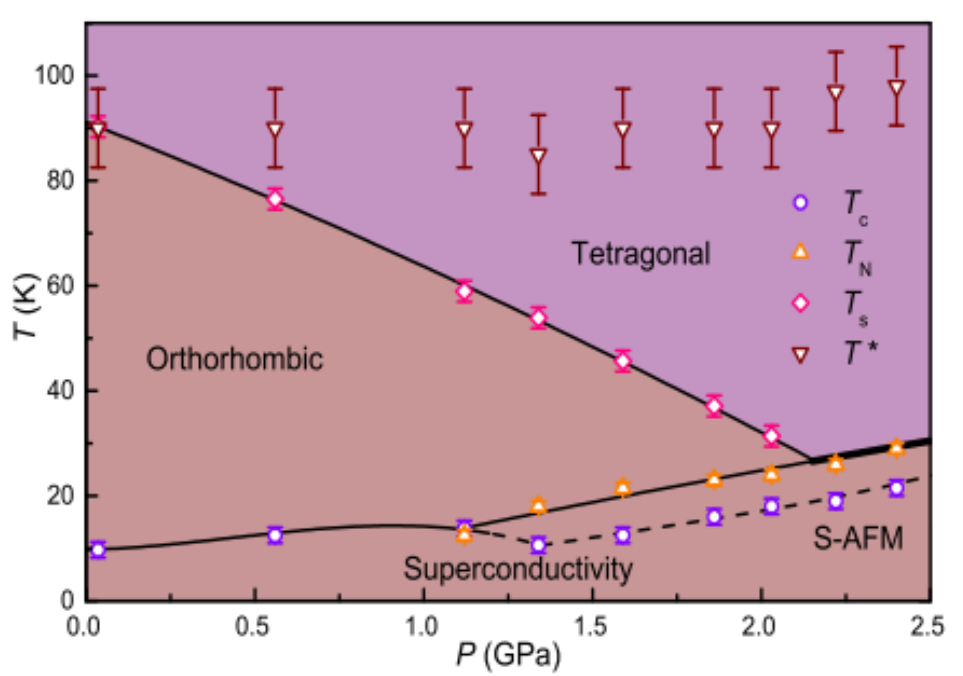


[1] M. Rotter, PRB 78, 020503 (2008)
[2] Y. Kamihara, . JACS , 130, 3296 (2008)

Structural and Magnetic Properties of FeSe

- Tetragonal (*P4/nmm*) to orthorhombic (*Cmme*), T_S = 90 K at ambient pressure
- No magnetic transition at ambient pressure, having T_C (8.5 K)
- Stripe-AFM above 2.4 GPa, no T_S

Phase diagram of FeSe [1]



[1] P. S. Wang, PRL 117, 237001 (2016)

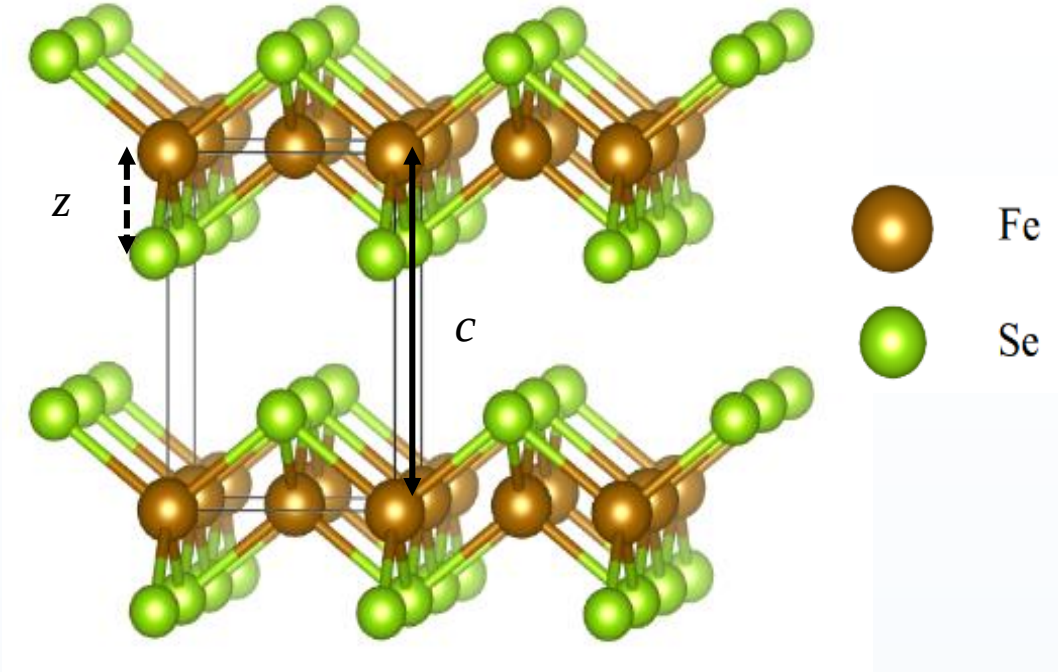
Purpose

- Study superconductivity of orthorhombic FeSe at ambient pressure with
  - Density Functional Theory (DFT)
  - Density Functional Perturbation Theory (DFPT)

- Calculate the superconducting temperature T_c of FeSe with McMillan's formula

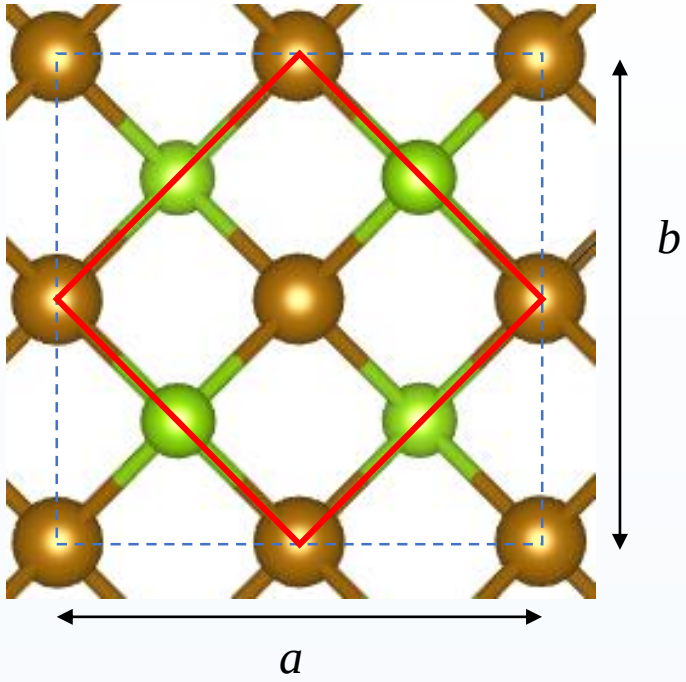
Structure of FeSe (low *T*)

- Base-centered orthorhombic
- *Cmme* (Group 67)
- Parameters:
  - *a, b, c*: lattice constants
  - *z*: chalcogen height (distance between Fe and Se layers)

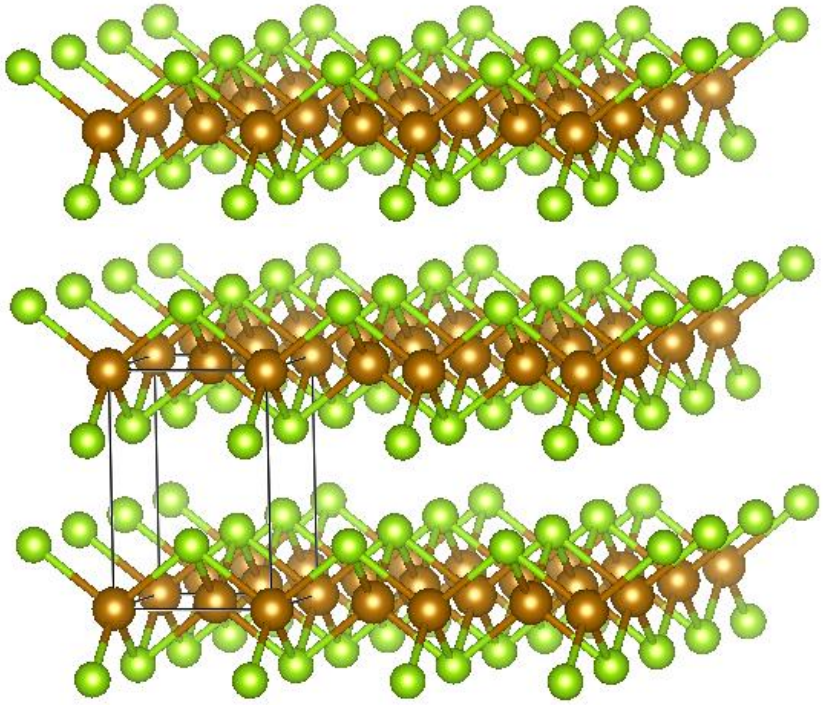


Structure of FeSe (low *T*)

Top view of the structure ($a \approx b$)



Side view of the structure



Structures for Calculations

- Structure A:
  - Lattice constants and atom positions from the experiments [1]
- Structure B:
  - Fully relax the lattice constants and atoms
- Structure C:
  - Relax only atoms, lattice constants fixed

- Relaxation method
  - PBE potential + Van der Waals correction with VASP
  - Non-magnetic Structure

[1] S. Margadonna, Y. Takabayashi, Chem. Commun. 5607 (2008)

Experimental Lattice Constants

High Temperature Data; *P4/nmm* (tetragonal structure)						
a (Å)		*c* (Å)	*z*	*P*	*T* (K)	
3.774		5.525	0.265	ambient	295	[1]
Low Temperature Data; *Cmme* (orthorhombic structure)						
a (Å)	*b* (Å)	*c* (Å)	*z*	*P*	*T* (K)	
5.308	**5.334**	**5.486**	**0.265**	**ambient**	**5**	[1]

[1] S. Margadonna, Y. Takabayashi, Chem. Commun. 5607 (2008)

Lattice Constants of the Structures

	a (Å)	*b* (Å)	*c* (Å)	*z*
Structure A [1]	5.3078	5.3342	5.486	0.2653
Structure B	5.1723	5.1726	5.4143	0.2563
Structure C	5.3078	5.3342	5.486	0.2436

Structure B and C are orthorhombic *(Cmme)*, same as Structure A

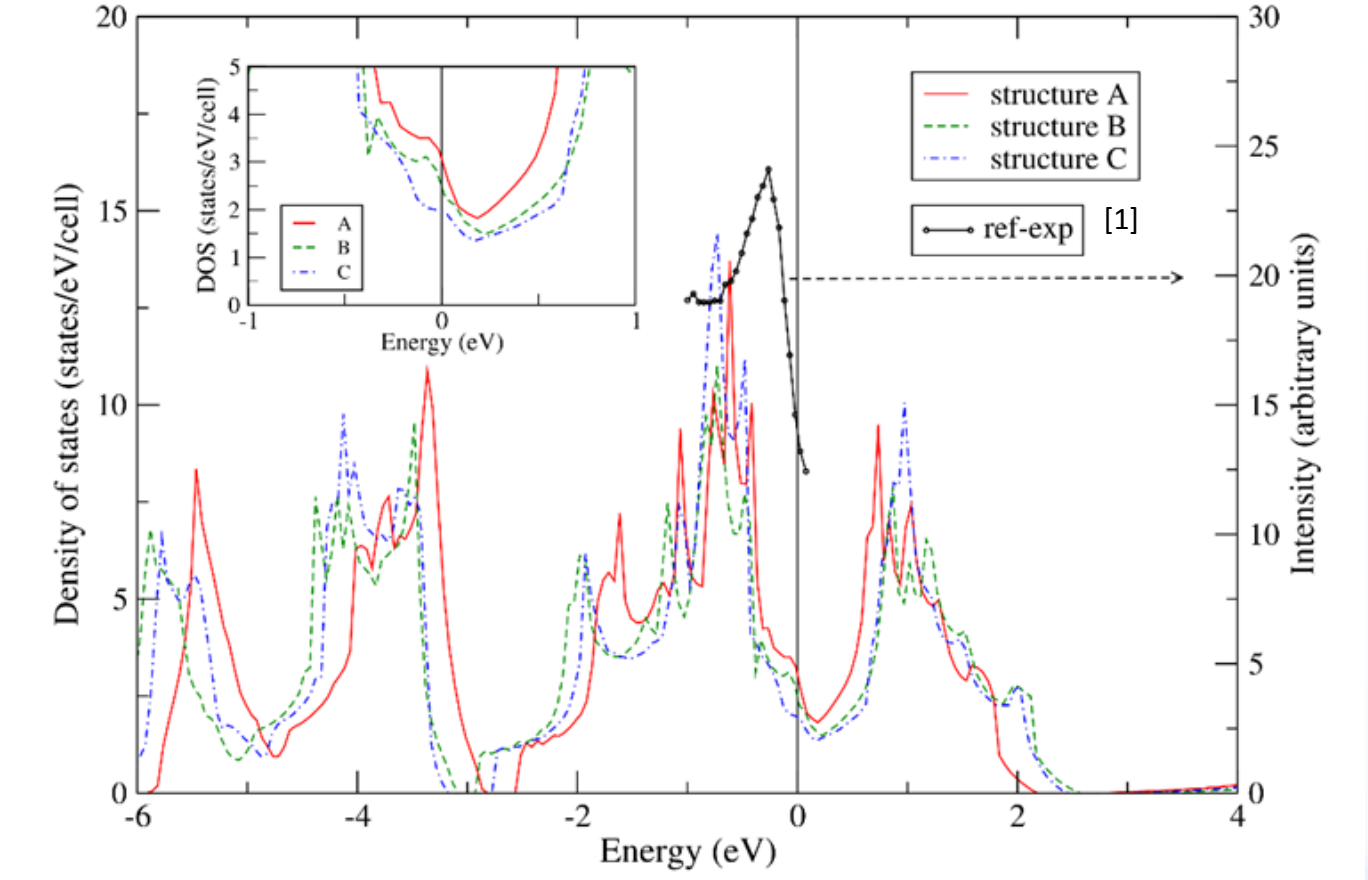
[1] S. Margadonna, Y. Takabayashi, Chem. Commun. 5607 (2008)

Calculation Method

- Calculation details
  - Quantum Espresso is used for both DFT and DFPT calculations.
  - PBE potential is used in the calculations with Van der Waals correction.

- Parameters for calculations
  - k-grid: 12x12x8
  - q-grid: 3x3x2
  - smearing: 0.04 Ry

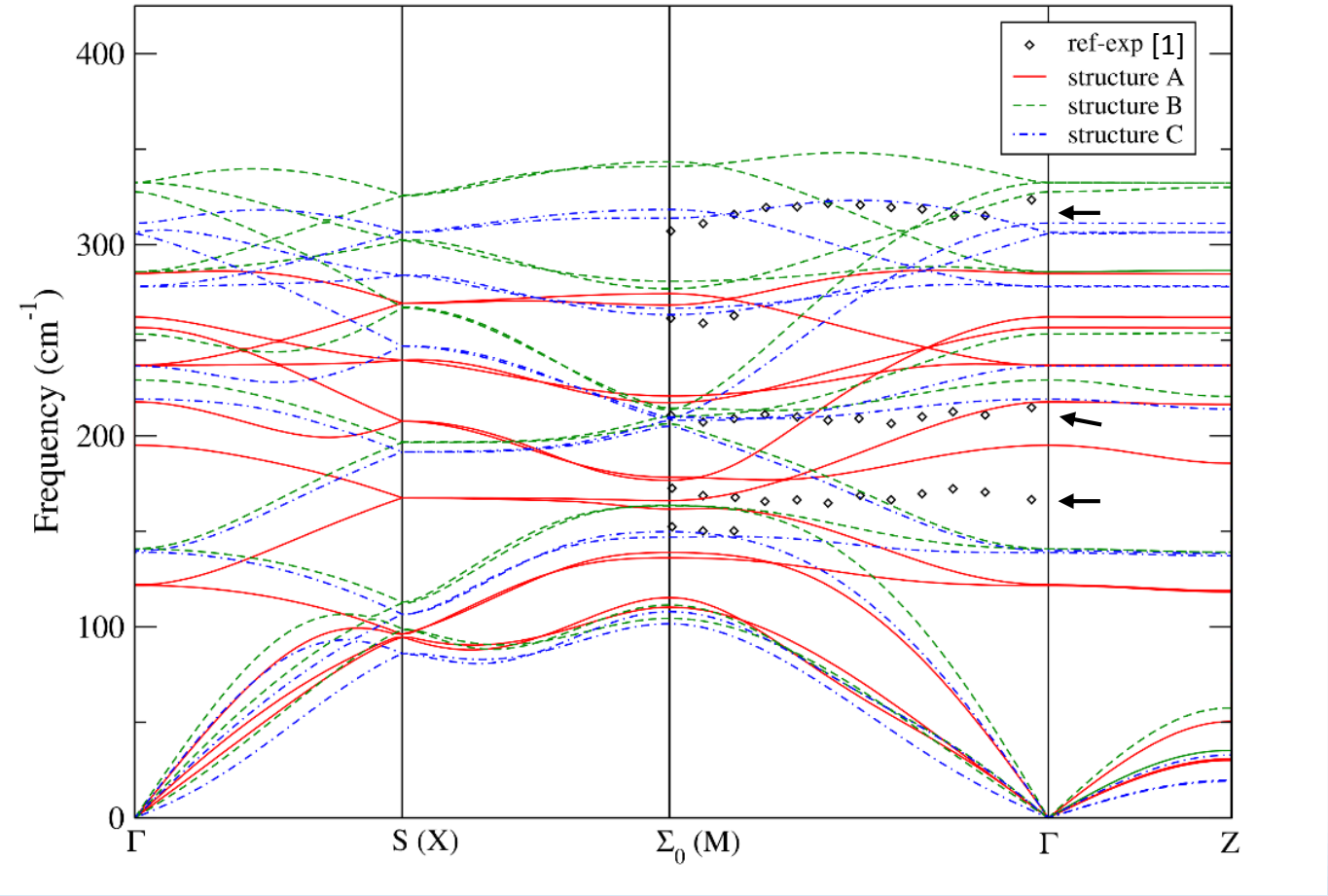
Density of States



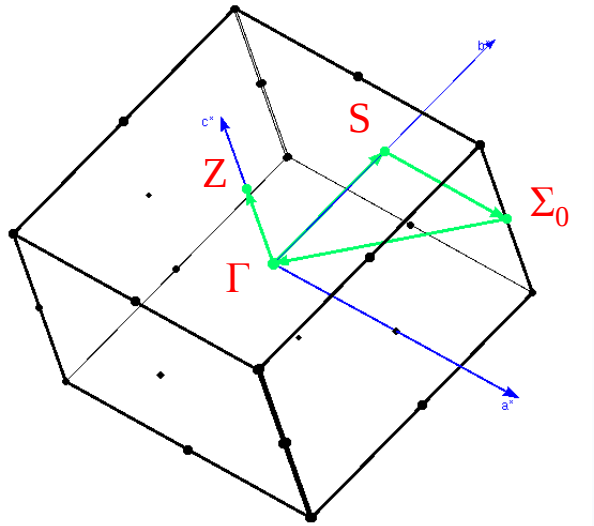
Structure	$D(\epsilon_f)$ (states/eV/cell)
A	3.0793
B	2.4762
C	1.9807

[1] R. Yoshida, J. Phys. Soc. Jpn., Vol. 78, No. 3 (2009)

Phonon Dispersion

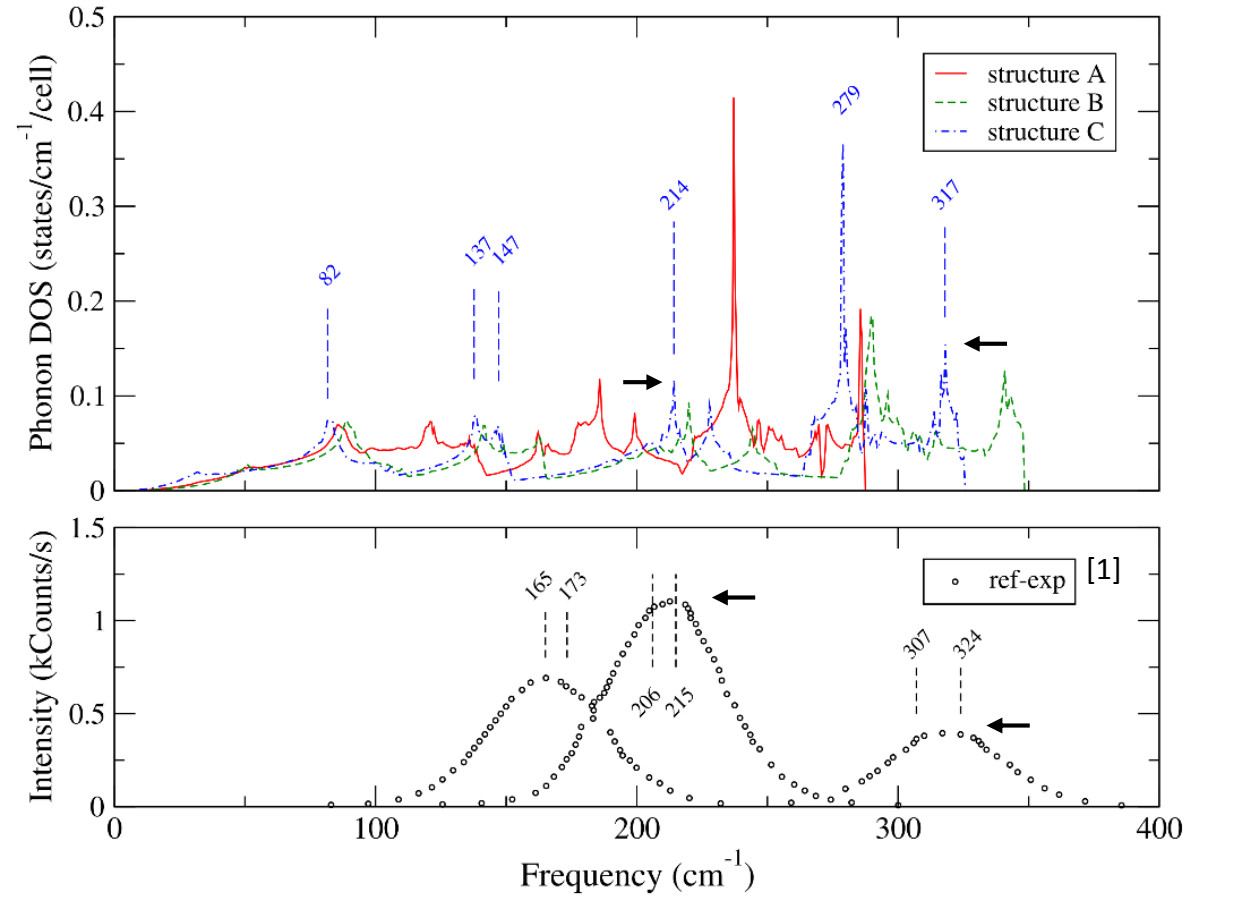


Brillouin zone of *Cmme* FeSe



[1] K. Zakeri, PRB 96, 094531 (2017)

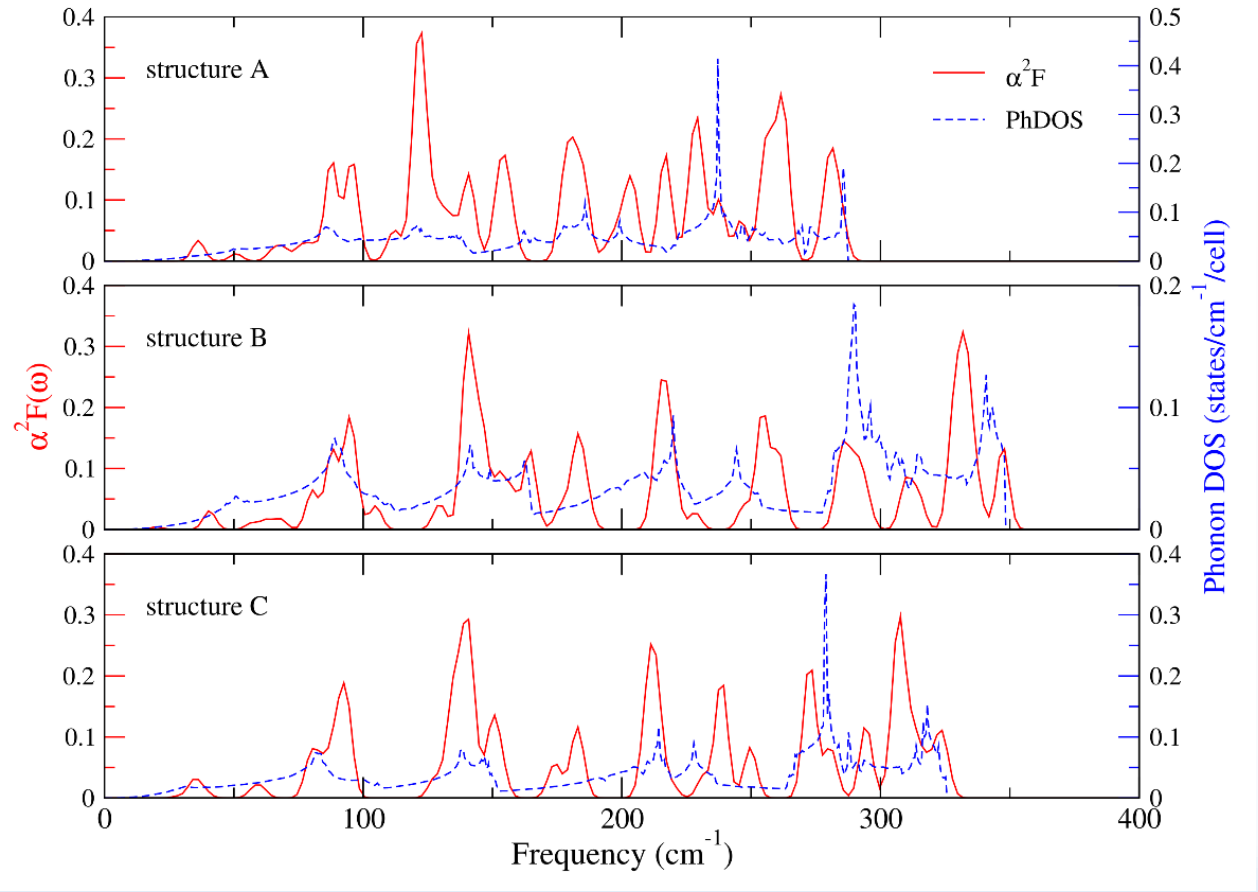
Phonon Density of States



- Calculated phonon DOS of three structures compared with three major modes measured in experiments [1].

[1] K. Zakeri, PRB 96, 094531 (2017)

Eliashberg function $\alpha^2F(\omega)$



- Calculated Eliashberg functions and phonon density of states of three structures

- $\lambda = 2\int_0^\infty \frac{1}{\omega}[\alpha^2F(\omega)]d\omega$

Calculation Results

	Structure	λ	$D(\epsilon_f)$ (states/eV/cell)	$\omega_{\log}$ (K)	T_c (K)
(1)	A	0.27	3.08	197.9	0.032
(2)	B	0.23	2.48	212.9	0.011
(3)	C	0.22	1.98	203.4	0.001
Thm [1]	C (no vdw)	0.17		162.6	$\ll 1$
Exp [2]					8.5

$\mu^* = 0.10$ for all calculations

$$T_c = \frac{\omega_{log}}{1.2} \exp\left[\frac{-1.04(1+\lambda)}{\lambda - \mu^*(1+0.62\lambda)}\right]$$

[1] A. Subedi, PRB 78, 134514 (2008)
[2] T. M. McQueen, PRB 79, 014522 (2009)

Is BCS still applicable to FeSe?

- $Ba_{1-x}K_xBiO_3$ (BKBO)
  - Measured T_C at about 30 K when x equal to 0.4[1].
  - Calculated T_C with DFPT method (< 6.1 K), much lower than experiments. [2]
- GW perturbation theory (from GW method)
  - Z. Li[2] calculated the T_C of $Ba_{0.6}K_{0.4}BiO_3$ within a range from 28.5 to 44.8 K
    - μ^* from 0.18 to 0.08
  - This was consistent with experiments.
- GWPT may possibly explain the superconductivity of FeSe.

[1] C. H. P. Wen, PRL 121, 117002 (2018)
[2] Z. Li, PRL 122, 186402 (2019)

Conclusion

- For structure C (atoms-only relaxed), its phonon dispersion matches the experiments better than other structures; however, its phonon DOS is not consistent with experiments, and it has the lowest electron-phonon coupling strength λ.
- The structure that follows experimental lattice constants (structure A) has the highest λ and $D(\epsilon_f)$, leading to highest T_C among all structures. Nevertheless, its T_C is still much lower than experiments (8.5 K) due to the small value of λ.
- As GW method solved the superconducting problem of $Ba_xK_{1-x}BiO_3$[1], it may solve the problem of FeSe.

[1] Zhenglu Li, PRL 122, 186402 (2019)

Acknowledgement

- Prof. Guang-Yu Guo, Department of Physics, NTU

Thank You for Your Attention!